

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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| In re Application of: | Thate et al. | Docket No.:       | 54129  |
| Application No.:      | 10/731,168   | Examiner:         | ONEILL |
| Filed:                | 12/10/2003   | Art Unit:         | 1795   |
| Customer No.:         | 26474        | Confirmation No.: | 5053   |

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For: Method of fabricating a membrane-electrode assembly

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Honorable Commissioner for Patents  
P.O. Box 1450  
Alexandria, Virginia 22313-1450

**DECLARATION UNDER 37 CFR 1.132**

I, Ömer Ünsal, a citizen of Germany, hereby declare:

I received a Ph.D. degree in Chemistry from Johannes-Gutenberg University in Mainz, Germany in 1999. I joined BASF SE on May 01, 2006, and I am currently a research Chemist. I have worked extensively in fuel cell research.

I have read and am familiar with the above-captioned patent application. I am familiar with the proceedings of the above-captioned patent application, including the Office action of July 28, 2009, which I have studied thoroughly. I have read and am familiar with US 6,346,182 to Bradley (hereinafter, "Bradley"). I would like to provide the following comments.

1. Bradley does not teach a membrane electrode assembly.

Independent claims 1, 6, and 13 of the present application relate to a method of fabricating a membrane-electrode assembly (MEA). Bradley is directed to a catalyst and to a method of producing the catalyst not to a method of fabricating an MEA for fuel

cells. The Examiner disagrees, arguing Bradley discloses a cellulose sheet used as a membrane between an anode and a cathode electrode. The Examiner's argument is entirely lacking in technical merit. A person having ordinary skill in the art would not agree with the Examiner's interpretation of the term "membrane electrode assembly for fuel cells." An MEA is not merely any membrane between an anode and a cathode. An MEA comprises a membrane and two gas diffusion electrodes. The membrane is highly proton conductive, but is an electrical insulator. The gas on the anode side of an MEA is separated by a membrane from the cathode air side. The gas tightness is very important for an MEA. Bradley does not teach a membrane-electrode assembly, but rather a laboratory procedure in which a cellulose sheet is used. The cellulose sheet would not allow for stable operation of a fuel cell. The cellulose sheet would not guarantee a separation of hydrogen and oxygen. Especially since the cellulose sheet of Bradley is highly porous and is not proton conductive, the cellulose sheet would not provide adequate separation between the hydrogen and oxygen compartment to function in a fuel cell. The high porosity of Bradley's cellulose sheet cannot be overlooked, because gas tightness is critical for an MEA.

2. Bradley's cellulose sheet is not a polymer-electrolyte membrane (PEM).

Claims 1, 6, and 13 of the present application further relate to a method of fabricating a membrane-electrode assembly (MEA), wherein the MEA comprises a PEM. Bradley's cellulose paper sheets (28) are not equivalent to a PEM. A PEM must be able to provide proton exchange. As already explained on page 1, lines 17 – 21 of the specification, in the case of a polymer-electrolyte membrane fuel cell (PEM fuel cell) the electrolyte used is a polymer membrane which conducts ions (especially  $H^+$  ions). The electrolyte has three functions. It establishes the ionic contact, prevents electrical contact, and additionally ensures that the gases fed to the electrodes are kept separate.

The Examiner has argued, Bradley's cellulose sheet is a polymer membrane, because "polymers are cellulose derivatives." This argument lacks technical merit, because the cellulose sheets are not proton exchange membranes. Cellulosic sheet material is not suitable for fuel-cell-arrangements. A person having ordinary skill in the

art would not expect Bradley's cellulose paper sheets to be useful as a PEM. Furthermore, Bradley provides no reason to assume that the cellulose sheets would be useful as a PEM. As discussed above, a thin paper sheet does not provide a separating function. Moreover, a porous and non-proton conductive paper sheet does not seal gases and provide ion transport current.

3. Bradley does not introduce ions of a catalytic component into the cellulose sheet or into an ionomer introduced into a reaction layer of the cellulose sheet.

Claims 1, 6, and 13 of the present application further relate to a method comprising A) introducing ions of the at least one catalytic component into the polymer-electrolyte membrane and/or into an ionomer introduced into the reaction layers. Bradley does not describe introducing ions of a catalytic component into a polymer-electrolyte membrane.

The Examiner seems to take two different positions on this issue. First, as can be seen, for example, on page 2 of the Office action, the Examiner argues introduction of catalyst ions occurs when the cellulose sheets are immersed into the liquid environment containing the catalyst. This theory lacks technical merit and is not supported by the disclosure of Bradley. Bradley disperses graphite particles onto one side of a cellulose paper sheet and then deposits Pd metal onto the graphite particles from a liquid solution containing  $\text{PdCl}_2$ . At column 14, lines 41 – 48,  $\text{Pd}^{2+}$  is only liberated from the solution upon creation of a sufficient electric field. The product of Bradley's procedure will not work as an MEA for fuel cell applications. As explained on page 1, lines 25 – 28 of the specification, the conversion of chemical into electrical energy takes place at the three-phase boundary of catalytically active sites (e.g. platinum), ion conductors (e.g. ion exchange polymers), electron conductors (e.g. graphite) and gases (e.g.  $\text{H}_2$  and  $\text{O}_2$ ). Ionic conductivity in the catalyst layer (three-phase boundary) is very important for a fuel cell, but is missing from Bradley.

The Examiner seems to present a second theory on page 9 by arguing Bradley introduces a catalyst component into the cellulose sheet when the catalytic substance is deposited onto the cellulose sheets. This second theory lacks technical merit. Based on

column 15, lines 20 – 25 of Bradley, Pd ions are introduced into the cellulose sheets when the Pd metal is deposited on the graphite. Again, ionic conductivity in the catalyst layer (three-phase boundary) is very important for a fuel cell, but is missing from Bradley.

4. Bradley does not describe variation of operating conditions during the deposition under fuel cell conditions.

Claim 3 of the present application relates to a method wherein a variation of operating conditions is effected during the deposition under fuel cell conditions. Bradley does not describe any variation in the operating conditions during the deposition. However, the Examiner argues, column 14, lines 50 – 54 of Bradley describes switching off the power supply. This argument neglects the phrase, “under fuel cell conditions.”

A person having ordinary skill in the art would understand “fuel cell conditions” to mean operation at a voltage of from 0 to 1.2 V, a current density of from 0 to 1.2 A/cm<sup>2</sup>, a temperature of from 25 to 200°C, and with hydrogen or reformat for anode and oxygen or air for cathode. Bradley does not describe any parameter concerning an operation stage of a fuel cell being under operation. According to Bradley, a mere deposition is disclosed.

I, Ömer Ünsal, further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued therefrom.

26<sup>th</sup> of October 2009  
Date

Ö. Ünsal  
Ömer Ünsal